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Customer No. 01333

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:

Kevin P. Klubek, et al

STABLE ORGANIC LIGHT-
EMITTING DEVICES USING
AMINOANTHRACENES

Group Art Unit: 1774

Examiner: Dawn L. Garrett

Serial No. US 10/753,249

Filed 08 January 2004

Commissioner for Patents
P.O. Box 1450
Alexandria, VA. 22313-1450

Sir:

DECLARATION UNDER RULE 132

I, the undersigned, Kevin P. Klubek, of Monroe County, New York, declare that:

I have received the degree of B.S. from the University of Buffalo for Medicinal Chemistry and am presently enrolled as a PhD candidate in Chemical Engineering at the University of Rochester.

Since June 1996, I have been engaged in research pertaining to Organic Light-Emitting Diode (OLED) technology focusing particularly on new material design and synthesis while also studying device efficiency and lifetime issues; I have many patents and publications on the subject and am considered an expert in this area;

I am a co-inventor in the above-captioned patent application;

I have reviewed the latest outstanding Office Action and any applicable cited references.

Amended claim 18 of the present application as currently amended is representative and requires a luminescent layer including the specified host and at least one dopant. The host of the luminescent layer is selected to include a solid organic material comprising a mixture of at least two components, wherein the first component is a non-

emissive organic compound containing the specified aminoanthracene, and the second component of the mixture contains an organic compound having a dipole moment larger than that of the first component. The dopant has a bandgap smaller than that of both the first and second components of the host and provides emission centers wherein the dopant produces blue, blue-green, green, green-yellow, or yellow.

An important feature of the subject matter of Amended Claim 18 is that it produces blue, blue-green, green, green-yellow, or yellow light with long lifetime and high efficiency. I believe this result to be unexpected.

At the request of Raymond L. Owens, an attorney of record, in accordance with the ordinary course of business of Eastman Kodak Company, I have supplied experimental data that supports the position that quite unexpected advantages were produced in accordance with the subject matter set forth in amended claim 18. The selection of the claimed dopant along with at least two host components having particular properties set forth in Amended Claim 18 produces results, which would provide an unpredictable and unexpected result.

The Examiner has rejected claims based on Oh et al. US003/0118866 that discloses two host materials that meet the requirements of Amended Claim 18 using a conventional red dopant. To show that that the selection of host materials and dopants is critical and not obvious, two very similar host materials that are utilized by Oh et al. for red devices are combined with a green dopant of the present invention. Oh et al. also uses Alq₃ as one of the hosts and an aminoanthracene as the second host. Oh et al. discloses a number of aminoanthracenes. The experimental data shown in the table below (IIA) uses the same generic type of aminoanthracene host as used by Oh et al. (Oh et al., Compound J-17, see paragraph 108 and paragraph 0125 of US2003/0118866) and is combined with Alq₃ and the green dopant DPQA of the present invention. For a comparison to the aminoanthracene of the type used by Oh et al., the aminoanthracene (AA1) was used in the new comparative experiments shown in Table IIA. AA1 is of the same generic type as disclosed by Oh et al. (J-17) and I believe, with reasonable technical certainty, would have similar performance. It is noteworthy that both AA1 and J-17 contain two diarylamine substituents.

| Compound ID | Structure |
|-------------|-----------|
| AA1 | |
| J-17 | |

Working Device Examples 1A-6A

The present invention and its advantages can be better appreciated by the following comparative examples. In these examples, the thickness of the organic layers and the doping concentration were controlled and measured in situ using calibrated thickness monitors (INFICON IC/5 Deposition Controller). The electroluminescence characteristics of all the fabricated devices were evaluated using a constant current source (KEITHLEY 2400 SourceMeter) and a photometer (PHOTO RESEARCH SpectraScan PR 650) at room temperature.

Example 1 (Comparative example)

The preparation of a conventional OLED is as follows: A ~1.1 mm thick glass substrate coated with a transparent ITO conductive layer was cleaned and dried using a commercial glass scrubber tool. The thickness of ITO is about 42 nm and the sheet resistance of the ITO is about 68 Ω /square. The ITO surface was subsequently treated with oxidative plasma to condition the surface as an anode. A layer of CF_x, 1 nm thick, was deposited on the clean ITO surface as the HIL by decomposing CHF₃ gas in an RF plasma treatment chamber. The substrate was then transferred into a vacuum deposition chamber (TROVATO MFG. INC) for deposition of all other layers on top of the substrate.

The following layers were deposited in the following sequence by evaporation from a heated boat under a vacuum of approximately 10^{-6} Torr: (1) a hole-transport layer, 75 nm thick, including NPB; (2) a luminescent layer, 37.5 nm thick, including Alq as the sole host material and 0.6% DPQA as the dopant; (3) an electron-transport layer, 37.5 nm thick, including AlQ₃; and (4) a cathode, approximately 2100 nm thick, including an alloy of magnesium and silver with a Mg:Ag volume ratio of about 10:0.5.

After the deposition of these layers, the device was transferred from the deposition chamber into a dry box (VAC Vacuum Atmosphere Company) for encapsulation.

The EL characteristics of this device were evaluated using a constant current source and a photometer. The drive voltage, luminance in cd/m^2 , EL efficiency in cd/A , and Commission Internationale de l'Eclairage (CIE) coordinates were all measured. Their values are shown in Table II. This device requires a drive voltage of 8.6 V to pass 20 mA/cm^2 . Its luminance is 1756 cd/m^2 and its luminance efficiency is 8.8 cd/A . The fabricated device was operated at 20 mA/cm^2 at room temperature for operational stability testing. The resulting luminance decay vs. operational time is shown in both FIG 4 and 5. After 300 hours operation, the luminance dropped by 13%.

Example 1A (Comparative example)

An OLED device similar to that of Comparative Example 1 was constructed, except that in the luminescent layer, Compound AA1 is the material for the first host component, Alq is the material for the second host component, and DPQA is the fluorescent dopant. The relative amounts of Compound AA1, Alq, and DPQA on a volume basis are in the ratio 5:94.4:0.6. The EL characteristics of this device are shown in Table IIA.

Example 2A (Comparative example)

An OLED device similar to that of Comparative Example 1 was constructed, except that in the luminescent layer, Compound AA1 is the material for the first host component, Alq is the material for the second host component, and DPQA is the fluorescent dopant. The relative amounts of Compound AA1, Alq, and DPQA on a volume basis are in the ratio 10:89.4:0.6. The EL characteristics of this device are shown in Table IIA.

Example 3A (Comparative example)

An OLED device similar to that of Comparative Example 1 was constructed, except that in the luminescent layer, Compound AA1 is the material for the first host component, Alq is the material for the second host component, and DPQA is the fluorescent dopant. The relative amounts of Compound AA1, Alq, and DPQA on a volume basis are in the ratio 25:74.4:0.6. The EL characteristics of this device are shown in Table IIA.

Example 4A (Comparative example)

An OLED device similar to that of Comparative Example 1 was constructed, except that in the luminescent layer, Compound AA1 is the material for the first host component, Alq is the material for the second host component, and DPQA is the fluorescent

dopant. The relative amounts of Compound AA1, Alq, and DPQA on a volume basis are in the ratio 50:49.4:0.6. The EL characteristics of this device are shown in Table IIA.

Example 5A (Comparative example)

An OLED device similar to that of Comparative Example 1 was constructed, except that in the luminescent layer, Compound AA1 is the material for the first host component, Alq is the material for the second host component, and DPQA is the fluorescent dopant. The relative amounts of Compound AA1, Alq, and DPQA on a volume basis are in the ratio 75:24.4:0.6. The EL characteristics of this device are shown in Table IIA.

Comparative examples 1A-5A contain a light emitting layer comprised of different concentrations of two host materials (AA1 and Alq) with a green dopant. Host compound AA1 is the equivalent as used by Oh et al. Looking at Table IIA, the lifetimes for all 5 devices have been improved compared to comparative device 1 that has only a single host, but this improvement results while paying a steep price in both efficiency and color purity. The efficiency drops for all devices and this decrease ranges from 8-16%, while the color shift ranges from 0.03-0.08 for the x coordinate and 0.02-0.06 for the y coordinate. The efficiency decreases and color shifts are both highly undesired and contrary to what is taught by the present invention.

Inventive examples shown in Table II (shown below and taken from the present specification) of the present invention are compared with comparative device 1 (as shown in Table I and also repeated below in Table II) having only a single host indicates that lifetime increases for all devices. Additionally, the efficiency increases, except for one data point where it remains about the same. All inventive devices retain or improve the color of the reference device. The inventive compounds of the present invention are non-emissive and this is verified by looking at the ElectroLuminescent (EL) spectra shown below in figure 1B. This figure shows that each of the inventive examples utilizing compound I as the first host component have EL spectra that are almost identical to that of the comparative reference device that has only a single host, Alq. In this manner, the color purity is not compromised. The small differences in the EL spectra for the inventive examples shown in Figure 1B, I attribute to solid-state solvation affects with respect to the dopant. In contrast, the EL spectra for the comparative devices 1A, 3A and 5A have all been shifted dramatically by addition of the aminoanthracene as taught by Oh et al. Even at the very low concentration of 5% (Comparative Example 1A), the shift is quite dramatic (the shoulder of the spectra has

increased in intensity). This indicates that emission is partly originating from AA1 and results in degradation of the color purity.

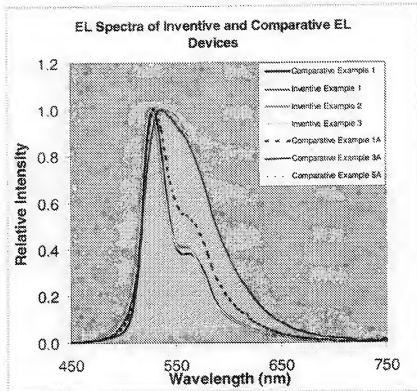
Table II of present invention (taken from paragraph 0122 of the present specification)

| Example | voltage (V) | luminance (cd/m ²) | efficiency (cd/A) | CIE _{x,y} | luminance loss after 300 hrs. |
|-----------------|-------------|--------------------------------|-------------------|--------------------|-------------------------------|
| 1 (comparative) | 8.6 | 1756 | 8.8 | 0.31, 0.65 | 13% |
| 2 (inventive) | 8.3 | 1857 | 9.3 | 0.31, 0.65 | 5% |
| 3 (inventive) | 7.6 | 2103 | 10.5 | 0.30, 0.65 | 4% |
| 4 (inventive) | 7.4 | 2019 | 10.1 | 0.30, 0.65 | 4% |
| 5 (inventive) | 7.7 | 1706 | 8.5 | 0.31, 0.65 | 7% |
| 6 (inventive) | 7.0 | 1880 | 9.4 | 0.31, 0.65 | 7% |
| 7 (inventive) | 6.9 | 1913 | 9.6 | 0.30, 0.64 | 8% |

Table IIA-new comparative data

| Example | voltage (V) | luminance (cd/m ²) | efficiency (cd/A) | CIE _{x,y} | luminance loss after 300 hrs. |
|------------------|-------------|--------------------------------|-------------------|--------------------|-------------------------------|
| 1 (comparative) | 8.6 | 1756 | 8.8 | 0.31, 0.65 | 13% |
| 1A (comparative) | 9.0 | 1515 | 7.58 | 0.34, 0.63 | 5% |
| 2A (comparative) | 8.8 | 1496 | 7.48 | 0.36, 0.61 | 6% |
| 3A (comparative) | 8.4 | 1629 | 8.14 | 0.39, 0.59 | 4% |
| 4A (comparative) | 7.5 | 1615 | 8.07 | 0.39, 0.59 | 5% |
| 5A (comparative) | 7.2 | 1480 | 7.40 | 0.38, 0.59 | 8% |

Figure 1B:



The selection of a coumarin dye that would produce blue, blue-green, green, green-yellow, or yellow light but not red light is well known in the art. This selection is clearly within the skill of one skilled in the art.

Therefore, with reasonable technical certainty, I have concluded that the subject matter of amended claim 18 is new, useful, and unobvious. The Oh et al. arrangement using an aminoanthracene with two diarylamine substituents is ineffective. The structure set forth in amended claim 18 with only one diarylamine substituent unexpectedly provides superior lifetime, excellent EL efficiency and excellent color purity that could not be predicted from the disclosure of Oh et al.

With reasonable technical certainty, it is clear to me and would also be clear to one skilled in the art after reading our disclosure that the materials covered by amended claim 18 would also provide unexpected results when producing blue, blue-green, green-yellow, or yellow light.

I declare further that all statements made herein of my own knowledge are true and all statements made on information and beliefs are believed to be true. These statements are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: _____

12/10/07

Kevin P. Klubek

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